(19) World Intellectual Property Organization International Bureau



(43) International Publication Date 1 February 2001 (01.02.2001)

PCT

(10) International Publication Number WO 01/07550 A1

(51) International Patent Classification7: C11D 3/28, D06M 13/355, 13/388

(21) International Application Number: PCT/EP00/05531

(22) International Filing Date: 15 June 2000 (15.06.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data: 9917450.0 23 July 1999 (23.07.1999) GB

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- (81) Designated States (national): AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

With international search report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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Fabric Treatment Composition and a Method of Treating Fabric

The present invention relates to a fabric treatment

composition comprising a novel colour protection treatment system. The present invention also relates to a method of treating fabric using the novel colour protection system.

The present invention further relates to the use of the novel colour protection system in the treatment of fabric.

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Background of the Invention

The present invention is concerned with fabric treatment that takes place after use of the fabric which may be referred to as fabric care. Such treatments include laundering, which uses detergents and/or fabric conditioner, and the application of non-detergent based fabric care products, such as spray-on products. Considerable attention has been focused on components for inclusion in laundry and non-detergent based care compositions, which components are specifically intended for deposition onto the fabric to protect the fabric and/or dye from damage. For example, these components may comprise photofading inhibitors and fluorescence inhibitors.

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Dye on fabric may be attacked by reactive radicals, thereby causing fading of the colour. The reactive radicals in question may be created by everyday chemicals (for example bleach or metal catalysed bleach) or by sunlight, maybe in combination with dirt or atmospheric pollutants.

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The present inventors have sought further colour and fabric protection systems which are suitable for incorporation into fabric treatment compositions for use in the laundering of fabrics and in fabric care.

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The present inventors have discovered that hindered amine light stabilisers can act as colour and fabric protection components in fabric treatment compositions for the laundering of fabrics and for fabric care.

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Hindered amine light stabilisers themselves are well known and used in the unrelated field of plastics manufacture.

Definition of the Invention

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The present invention accordingly provides a fabric treatment composition for use in a method of laundry fabric treatment or non-detergent based fabric care treatment, comprising a hindered amine light stabiliser.

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The present invention further provides a method for the treatment of fabric, comprising treating the fabric in a method of laundry fabric treatment or non-detergent based fabric care treatment with a hindered amine light

25 stabiliser.

The present invention further provides use of a hindered amine light stabiliser in a method of laundry fabric treatment or non-detergent based fabric care treatment.

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Detailed Description of the Invention

Hindered Amine Light Stabilisers

Hindered amine light stabilisers are known as oxidation inhibition materials particularly for use with thermoplastics. They are described generally in "Oxidation Inhibition in Organic Materials" Vol. II, Pospisil and Klemchuk, 1990, Florida, pages 2-25.

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They are molecules which include the tetramethyl piperidine moiety in their structure:

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wherein R1 and R2 comprise suitable substituents.
R2 is suitably hydrogen, alkyl or alkoxy.

In a preferred embodiment, R_2 is an oxygen atom with an unpaired electron on it, forming a nitroxide radical with the N atom.

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In the hindered amine light stabilisers set out in US 5874493, R_1 is hydrogen, hydroxyl, low molecular weight alkyl (for example C_1 - C_8), low molecular weight alkoxy (eg C_1 - C_8) or acyl and R_2 is:

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 $-R_3-OC = NH-R_4$,

wherein R_3 is oxygen, NH- or $N(C_1-4-alkyl)$ - and R_2 is mono or bi-nuclear aromatic group which optionally bears substituents.

Hindered amine light stabilisers are typically available under the trade mark Tinuvin .

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It has been surprisingly found that levels of hindered amine light stabiliser as low as 0.01-2% on-weight-fabric are sufficient to counteract the effect of reactive radicals, particularly leading to a reduction in damage to dye.

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Particular suitable hindered amine light stabilisers are selected from the group consisting of 4-hydroxy-2,2,6,6-tetramethyl piperidine, bis(2,2,6,-tetramethyl piperidyl)sebacate (Tinuvin 770, trade mark), bis(1-methyl-2,2,2,6,-tetramethylpiperidyl)sebacate (Tinuvin 765, trade mark), decanedioic acid, bis(2,2,6,6-tetramethyl-1-(octyloxy)-4-piperidyl)ester (Tinuvin 123, trade mark), 4-hydroxy-2,2,4,4-tetramethyl piperinyloxy, and mixtures thereof.

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Hindered amine light stabilisers are suitably present at levels in the range 0.1-5% by weight in compositions according to the present invention.

- 5 -

Fabric Treatment Compositions

The present invention is suitable for use in fabric wash compositions, fabric conditioning compositions and compositions for both washing and conditioning fabrics (so-called through the wash conditioner compositions). The present invention can also be applied to industrial and domestic non-detergent based fabric care compositions, for example spray-on compositions.

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Fabric Wash Compositions

Fabric wash compositions according to the present invention comprise a fabric wash detergent material selected from non-soap anionic surfactant, nonionic surfactants, soap, amphoteric surfactants, zwitterionic surfactants and mixtures thereof.

Suitable anionic surfactants are well known to the person skilled in the art and include alkyl benzene sulphonate, primary and secondary alkyl sulphates, particularly C₈-C₁₅ primary alkyl sulphates; alkyl ether sulphates; olefin sulphonates; alkyl xylene sulphonates, dialkyl sulphosuccinates; ether carboxylates; isethionates; sarcosinates; fatty acid ester sulphonates and mixtures thereof. The sodium salts are generally preferred.

Nonionic surfactants are also well known to the person skilled in the art and include primary and secondary alcohol ethoxylates, especially C_8 - C_{20} aliphatic alcohol ethoxylated with an average of from 1 to 20 moles of ethylene oxide per

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mole of alcohol, and more especially the C₁₀-C₁₅ primary and secondary aliphatic alcohols ethoxylated with an average of from 1 to 10 moles of ethylene oxide per mole of alcohol.

Non-ethoxylated nonionic surfactants include alkyl polyglycosides, glycerol monoethers and polyhydroxy amides (glucamide). Mixtures of nonionic surfactant may be used.

Detergent compositions suitable for use in domestic or industrial automatic fabric washing machines generally contain anionic non-soap surfactant or nonionic surfactant, or combinations of the two in suitable ratio, as will be known to the person skilled in the art, optionally together with soap.

- Many suitable detergent-active compounds are available and fully described in the literature, for example in Surface-Active Agents and Detergents , Volumes I and II, by Schwartz, Perry & Berch.
- Anionic surfactant is suitably present at a level of from 5 wt% to 50 wt%, preferably 10 wt%-40 wt% based on the fabric treatment composition. Nonionic surfactant is suitably present at a level of 1-20 wt%, preferably 5-15 wt%.
- The total amount of surfactant present will depend upon the intended end use and may be as high as 60 wt% for example in a composition for washing fabrics by hand. In compositions for machine washing of fabric, an amount of from 5 to 40 wt% is generally appropriate.

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Detergency Builder

The detergent compositions of the invention will generally also contain one or more detergency builders. amount of detergency builder in the compositions will suitably range from 5 to 80 wt%, preferably from 10 to 60 wt%.

Inorganic builders that may be present include sodium carbonate, if desired in combination with a crystallisation 10 seed for calcium carbonate, as disclosed in GB 1 437 950 (Unilever); crystalline and amorphous aluminosilicates, for example, zeolites as disclosed in GB 1 473 202 (Henkel) and mixed crystalline/amorphous aluminosilicates as disclosed in GB 1 470 250 (Procter & Gamble); and layered silicates as 15 disclosed in EP 164 514B (Hoechst). Inorganic phosphate builders, for example, sodium orthophosphate, pyrophosphate and tripolyphosphate are also suitable for use with this invention.

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The detergent compositions of the invention preferably contain an alkali metal, preferably sodium, aluminosilicate builder. Sodium aluminosilicates may generally be incorporated in amounts of from 10 to 70% by weight (anhydrous basis), preferably from 25 to 50 wt%.

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The alkali metal aluminosilicate may be either crystalline or amorphous or mixtures thereof, having the general formula:

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These materials contain some bound water and are required to have a calcium ion exchange capacity of at least 50 mg CaO/g. The preferred sodium aluminosilicates contain 1.5- $3.5 \, \mathrm{SiO_2}$ units (in the formula above). Both the amorphous and the crystalline materials can be prepared readily be reaction between sodium silicate and sodium aluminate, as amply described in the literature.

Suitable crystalline sodium aluminosilicate ion-exchange
detergency builders are described, for example, in GB 1 429
143 (Procter & Gamble). The preferred sodium
aluminosilicates of this type are the well-known
commercially available zeolites A and X, and mixtures
thereof.

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The zeolite may be the commercially available zeolite 4A now widely used in laundry detergent powders. The zeolite builder incorporated in the compositions of the invention may be maximum aluminium zeolite P (zeolite MAP) as

20 described and claimed in EP 384 070A (Unilever). Zeolite MAP is defined as an alkali metal aluminosilicate of the zeolite P type having a silicon to aluminium ratio not exceeding 1.33, preferably within the range of from 0.90 to 1.33, and more preferably within the range of from 0.90 to 1.20.

Especially preferred is zeolite MAP having a silicon to aluminium ratio not exceeding 1.07, more preferably about 1.00. The calcium binding capacity of zeolite MAP is generally at least 150 mg CaO per g of anhydrous material.

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Organic builders that may be present include polycarboxylate polymers such as polyacrylates, acrylic/maleic copolymers, and acrylic phosphinates; monomeric polycarboxylates such as citrates, gluconates, oxydisuccinates, glycerol mono-, diand trisuccinates, carboyxmethyloxysuccinates, carboxymethyloxymalonates, dipicolinates, hydroyxethyliminodiacetates, alkyl- and alkenylmalonates and succinates,; and sulphonated fatty acid salts. This list is not intended to be exhaustive.

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Especially preferred organic builders are citrates, suitable used in amounts of from 5 to 30 wt%, preferably from 10 to 25 wt%; and acrylic polymers, more especially acrylic/maleic copolymers, suitably used in amounts of from 0.5 to 15 wt%, preferably from 1 to 10 wt%.

Builders, both inorganic and organic, are preferably present in alkali metal salt, especially sodium salt, form.

20 Bleach Components

Detergent compositions according to the invention may also suitably contain a peroxy bleach system for example, inorganic persalts or organic peroxyacids, capable of yielding hydrogen peroxide in aqueous solution.

Suitable peroxy bleach compounds include organic peroxides such as urea peroxide, and inorganic persalts such as the alkali metal perborates, percarbonates, perphosphates, persilicates and persulphates. Preferred inorganic persalts

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are sodium perborate monohydrate and tetrahydrate, and sodium percarbonate.

Especially preferred is sodium percarbonate having a protective coating against destabilisation by moisture. Sodium percarbonate having a protective coating comprising sodium metaborate and sodium silicate is disclosed in GB 2 123 044B (Kao).

10 Fabric Softening Composition

The fabric treatment composition of the present invention may be a fabric conditioning composition or it may comprise fabric conditioner.

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Fabric Softening Compound

The fabric softening compound is preferably a cationic nonionic or anionic fabric softening compound.

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The fabric softening compound may be a quaternary ammonium material comprising a polar head group and one or two alkyl or alkenyl chains. The fabric softening compound may also be a nonionic fabric softening compound such as a fabric softening oil, a fabric softening silicone composition or a fabric softening ester composition such as sugar esters.

Particularly preferably, the fabric softening compound has two long chain alkyl or alkenyl chains with an average chain length greater than C₁₄, more preferably each chain

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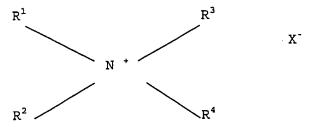
has an average chain length greater than C_{14} , more preferably at least 50% of each long chain alkyl or alkenyl group has a chain length of C_{18} .

5 It is preferred if the long chain alkyl or alkenyl groups of the fabric softening compound are predominantly linear.

It is highly preferred if the fabric softening compounds are substantially water-insoluble. Substantially insoluble fabric softening compounds in the context of this invention are defined as fabric softening compounds having a solubility less than 1 x 10⁻³ wt% in demineralised water at 20°C, preferably the fabric softening compounds have a solubility less than 1 x 10⁻⁴, most preferably the fabric softening compounds have a solubility at 20°C in demineralised water from 1 x 10⁻³ to 1 x 10⁻⁶.

Well known species of substantially water-insoluble quaternary ammonium compounds having the formula:

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wherein R^1 and R^2 represent hydrocarbyl groups having from 12 to 24 carbon atoms; R^3 and R^4 represent hydrocarbyl groups containing 1 to 4 carbon atoms; and X is an anion,

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preferably selected from halide, methyl sulphate and ethyl sulphate groups are preferred.

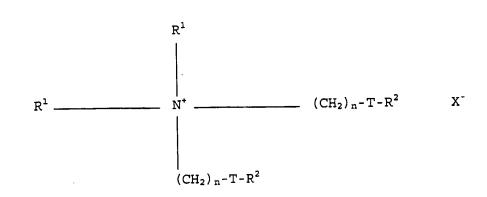
Representative examples of these quaternary softeners
include di(tallow alkyl) dimethyl ammonium methyl
sulphate; dihexadecyl dimethyl ammonium chloride;
di(hydrogenated tallow alkyl) dimethyl ammonium chloride;
dioctadecyl dimethyl ammonium chloride; di(hydrogenated
tallow alkyl) dimethyl ammonium methyl sulphate;
dihexadecyl diethyl ammonium chloride; di(coconut alkyl)
dimethyl ammonium chloride, ditallow alkyl dimethyl
ammonium chloride and di(hydrogenated tallow alkyl)
dimethyl ammonium chloride (Arquad 2HT Trade Mark).

Other preferred softeners contain esters or amide links, for example those available under the trade names Accosoft 580, Varisoft 222, and Stepantex.

Particularly preferred fabric softening compounds are

water-insoluble quaternary ammonium materials which
comprise a compound having two C₁₂₋₁₈ alkyl or alkenyl
groups connected to the molecule via at least one ester
link. It is more preferred if the quaternary ammonium
material has two ester links present. The preferred
ester-linked quaternary ammonium material for use in the
invention can be represented by the formula:

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wherein each R^1 group is independently selected from C_{1-4} alkyl, hydroxyalkyl or C_{2-4} alkenyl groups; and wherein each R^2 group is independently selected from C_{8-28} alkyl or alkenyl groups;

T is -C-O- or -O-C-; X is any suitable anion and n is an integer from 0-5. Particularly preferred is di(ethyl ester) dimethyl ammonium chloride (DEEDMAC).

20 A second preferred type of quaternary ammonium material can be represented by the formula:

wherein R_1 , n, $X^{\scriptscriptstyle \text{I}}$ and R_2 are as defined above.

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It is advantageous for environmental reasons if the quaternary ammonium material is biologically degradable.

Preferred materials of this class such as 1,2 bis[hardened tallowoyloxy]-3-trimethylammonium propane chloride and their methods of preparation are, for example, described in US 4 137 180 (Lever Brothers). Preferably these materials comprise small amounts of the corresponding monoester as described in US 4 137 180 for example 1-hardened tallowoyloxy-2-hydroxy trimethylammonium propane chloride.

The fabric softening agent may also be a polyol ester quat (PEO) as described in EP 0 638 639 (Akzo).

15 Other Ingredients

The compositions of the invention can also contain one or more optional ingredients, selected from pH buffering agents, perfume carriers, fluorescers, colorants, hydrotropes, antifoaming agents, antiredeposition agents, enzymes, optical brightening agents, opacifiers, antishrinking agents, anti-wrinkle agents, anti-spotting agents, germicides, fungicides, anti-corrosion agents, drape

imparting agents, antistatic agents and ironing aids.

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The present invention may be in the form of a dilute or concentrated aqueous solution or suspension, for example as described in WO 97/15651, WO 95/27769. Alternatively, the fabric softening composition may be in the form of a powder for use in the rinse cycle of an automatic washing machine. Alternatively, the fabric softening composition may be in

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the form of a sheet comprising fabric conditioning compositions for use in a tumble dryer, for example as disclosed in WO 95/27777. Alternatively, the fabric conditioning composition may be in the form of a substantially non-aqueous concentrate as described in international patent application to PCT/EP99/00497.

Fabric wash detergent compositions according to the present invention may further include through the wash softening material, such as cationic fabric softener.

Non-Detergent Based Fabric Care Products

The present invention can also be used in non-detergent

based fabric care products. For example, the product may comprise the hindered amine light stabiliser as the principal ingredient. For example, non-detergent based compositions may comprise solutions of the hindered amine light stabiliser of the present invention in a suitable solvent, such as isopropanol, alcohol etc. The compositions may comprise aerosol or spray-on compositions. They may be in the form of sticks, bars, dab-on compositions, for example absorbed into sponges for application to the surface etc.

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Such non-detergent based fabric care products are products which are not primarily intended for removal of soil or stains or for fabric softening. They may, however, include surfactant for other reasons, for example stabilising components, encouraging spread or deposition of the products onto fabric, viscosity control etc. Suitably, the

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surfactant level is below 5% by weight, more preferably below 1% by weight.

The present invention will be further described by way of example only with reference to the following examples.

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Examples

All quantities are in percent by weight or parts by weight unless indicated otherwise.

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The following hindered amine light stabilisers are used in the present examples. They are available commmercially from Ciba Specialty Chemicals:

15 HTMP = 4-hydroxy-2,2,6,6-tetramethyl piperidine.

Tinuvin 770 = bis(2,2,6,6-tetramethyl piperidyl)sebacate.

Tinuvin 765 = bis(1-methyl,2,2,6,6-tetramethyl
20 piperidyl)sebacate.

Tinuvin 123 = decanedioic acid, bis(2,2,6,6-tetramethyl-1-(octyloxy)-4-piperidyl ester.

25 Reflectance is measured using a Spectraflash SF60 reflectometer (trade mark). The weathermeter is an Atlas S3000 (trade mark).

Example 1

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A range of dyed cotton fabric (printed on one side) was purchased from Veebee Tech, (Harrogate, England). The dyes used are listed in Table 1.

Benzoyl peroxide was then applied to each of the test cloths from a solution of benzoyl peroxide in ethanol, to give 0.09% on-weight-fabric benzoyl peroxide.

Secondly, hindered amine light stabilisers were deposited onto the test cloths from ethanol solution at the levels indicated in Table 1. For each dye, a control cloth was produced which contained no hindered amine light stabiliser.

Subsequently, the controlled test cloth and the treated test cloths were each placed in boiling water at 100°C for 60 seconds. Under these conditions, benzoyl peroxide decomposes to give reactive radicals which will attack dye.

In order to determine the extent of dye damage due to the formation of reactive radicals by benzoyl peroxide, the change in reflectance before and after the boiling treatment for each cloth was determined and measured in CIELAB ΔE units. Table 1 shows the CIELAB ΔE units for the control cloths, cloths treated with 0.25% on-weight-fabric Tinuvin 770 (trade mark) and cloths treated with 0.69% on-weight-fabric of Tinuvin 765 (trade mark).

Table 1

		CIELAB AE			
Dye type	wt% dye	Control	Cloth treated	Cloth	
	on	cloth	with 0.25% owf.	treated with	
	weight	1	Tinuvin 770	0.69% owf.	
	fabric		(trade mark)	Tinuvin 765	
•				(trade mark)	
Procion					
Scarlet HE-3G	0.05	3.47	2.80	1.33	
Crimson HE-XL	0.05	2.04	1.77	1.05	
Yellow HE-6G	0.1	0.66	0.63	0.64	
Red HE-XL	0.05	3.97	2.45	1.61	
Blue HE-XL	0.2	34.16	34.02	30.07	
Turquoise H-A	0.5	33.18	23.57	5.61	
Navy HE-XL	0.1	4.84	3.04	1.61	
Remazol					
Red RB	0.1	1.12	0.71	0.99	
Brilliant Red	0.2	0.66	0.66	0.48	
RBS					
Orange FR	0.2	1.38	0.58	1.12	
Navy GG	0.2	6.99	4.81	4.23	
Turquoise G	0.2	26.06	24.45	13.57	
Black B	0.2	6.01	4.79	4.03	

It can be seen that for a wide range of dyes, hindered amine
light stabilisers at relatively low on-weight-fabric levels
can reduce dye damage due to reactive radicals.

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Example 2

Dyed test cloths having the dyes listed in Table 2 were obtained as in Example 1. Control dyed test cloths were taken and received no further treatment. The other dyed test cloths were treated with hindered amine light stabiliser by depositing the hindered amine light stabiliser from ethanolic solution to give the on-weight-fabric levels indicated in Table 2.

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The control cloths and cloths treated with hindered amine light stabiliser were subjected to simulated sunlight in an Atlas S3000 Weatherometer (trade mark) at a power of 0.35 W/m^2 at 340 nm. The change in colour intenstiy was measured using a Spectraflash SF600 Reflectometer (trade mark) and expressed in CIELAB ΔE units. The results are reported in Table 2.

Table 2

Dye	Treatment	ΔΕ	Hours irradiation
sulphol	none	2.4	4
orange			
	0.15% owf Tinuvin 770	0.1	4
	(trade mark)		
	0.15% owf Tinuvin 765	1.6	4
	(trade mark)	_	
	none	4.95	9
	0.16% HTMP	1.97	9
Blue HE-XL	none	3.22	15
	0.06% Tinuvin 123 (trade	2.40	15
	mark)		
	0.17% HTMP	1.32	15

It can be seen that hindered amine light stabilisers

according to the present invention reduce simulated sunlight
damage to dye under a wide range of circumstances.

Example 3

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- 10 The following fabric rinse conditioner compositions were prepared:
 - (a) HEQ 4% by weight
 Minors 0.05% by weight
 water to 100% by weight
 - (b) HEQ 4% by weight

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Tinuvin 765 (trade mark) 5.3% by weight.

Minors - 0.5% by weight

Water to 100% by weight

- 5 (c) HEQ 4% by weight
 Tinuvin 765 (trade mark) 1.5% by weight
 Minors 0.5% by weight
 Water to 100% by weight
- 10 Each conditioner composition was prepared by dispersing the components in water with vigorous mixing.
 - HEQ = 1,2 bis [hardened tallowoyloxy]-3-trimethyl ammonium
 propane chloride, ex Hoechst.

Dyed cloths were obtained as in Example 1.

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For each colour of dye, 2.4g of dyed cloth was taken and treated with 100g of demineralised water into which 1g of 20 rinse conditioner (a), (b) or (c) had been dispersed.

After a minute of immersion, the cloth was removed and dried. Subsequently, 0.12% on-weight-fabric benzoyl peroxide was padded onto the cloth from an ethanol solution. The ethanol was evaported. The cloth was then placed in boiling demineralised water for 1 minute, removed, and dried. The colour of each cloth was measured before and after the treatment with benzoyl peroxide. The change in colour measured in CIELAB ΔE units was measured. The

30 results were shown in Table 3.

Table 3

	ΔΕ			
Dye	(a)	(b)	(c)	
procion red HE-7B	2.6	1.8	2.1	
procion scarlet HE-3G	2.0	1.3	1.7	
procion crimson HE-XL	1.6	1.0	1.5	
procion turquoise H-A	41.2	22.0	37.1	
procion navy HE-XL	3.9	3.3	3.6	

It can be seen that the rinse conditioner compositions (b)

and (c) comprising hindered amine light stabilisers result
in lower changes in colour than the control composition (a).

Thus, hindered amine light stabilisers are shown to be
deposited from rinse conditioner compositions and to give
protection to fabric colour.

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CLAIMS:

1. A fabric treatment composition for use in a method of laundry fabric treatment or non-detergent based fabric care treatment, comprising a hindered amine light stabiliser.

2. The composition of claim 1, being a fabric conditioner composition, the composition further comprising fabric conditioner.

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- 3. The composition of claim 2, being a rinse conditioner.
- 4. The composition of claim 1, being a fabric wash composition, the composition further comprising fabric wash detergent.
- 5. The composition of claim 4, wherein the fabric wash detergent is selected from non-soap anionic, nonionic, amphoteric and zwitterionic surfactants and mixtures thereof.
- 6. The composition of claims 4 or 5, further comprising a builder material.
- 7. The composition of any preceding claim, wherein the hindered amine light stabiliser includes a tetramethyl piperidine moiety.
- 8. A method for the treatment of fabric, comprising30 treating the fabric with a hindered amine light stabiliser

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in a method of laundry fabric treatment or non-detergent based fabric care treatment.

- 9. A method according to claim 8, wherein the treatment of fabric takes place during a fabric washing step.
 - 10. A method according to claim 8, wherein the treatment takes place during a fabric rinsing step, in the presence of fabric conditioner.

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11. Use of a hindered amine light stabiliser to prevent damage to dyed fabric due to reactive radicals, comprising treating the dyed fabric with a composition comprising the hindered amine light stabiliser as part of a laundry fabric treatment or non-detergent based fabric care process.

INTERNATIONAL SEARCH REPORT

PCT/EP 00/05531

A. CLASSIFICATION OF SUBJECT MATTER IPC 7 C11D3/28 D06M D06M13/355 D06M13/388 According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) C11D D06M IPC 7 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, WPI Data, PAJ C. DOCUMENTS CONSIDERED TO BE RELEVANT Relevant to claim No. Citation of document, with indication, where appropriate, of the relevant passages EP 0 315 204 A (LION CORP ; SANKYO CO (JP)) 1,4-9,11X 10 May 1989 (1989-05-10) claims 1, 3, 4.7-14examples page 2, line 49 -page 3, line 11 page 13, line 29 - line 39 WO 97 33034 A (RHONE-POULENC CHIMIE) 1,7,8,11 X 12 September 1997 (1997-09-12) claims 1-10 examples 2,3,10 A Patent family members are listed in annex. Further documents are listed in the continuation of box C. Special categories of cited documents: T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the "A" document defining the general state of the art which is not considered to be of particular relevance invention "E" earlier document but published on or after the international "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "&" document member of the same patent family Date of mailing of the international search report Date of the actual completion of the international search 13/09/2000 5 September 2000 Authorized officer Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo ni, Neys, P Fax: (+31-70) 340-3016

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i. national Application No PCT/EP 00/05531

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